

History of maltitol crystallization

1. Hirao et al., in US 4,408,041 (filed with IDS), assigned to Hayashibara, with a priority date of Feb. 1981, were the first to describe anhydrous maltitol crystals.

They start with a maltitol syrup with a high content in maltitol, hence a low content in polyols of higher degree of polymerization (DP3, DP \geq 4), in particular maltotriitol.

More precisely, in the description Col. 2 lines 57-60, the maltitol solution is said to contain up to about 98.5% maltitol. In example 2, the sugar mixture which is crystallized contains 92.2% maltitol, 0.8% sorbitol, 4.6% DP3, and 2.4% DP \geq 4. When the maltitol content is closer to 98.5%, the maltotriitol content will very probably be less than 1%.

Following crystallization out of water, the crystals are said to be "*colorless, odorless, transparent crystals (micro-crystals in lump white)*", see Col. 3 lines 53-54.

In the article corresponding to the patent in *Carbohydrate Research*, published in 1982 (filed with IDS), the crystals are described as being "*colorless, prism-like crystals*", see page 164, X-Ray crystallographic analysis.

Hirao et al., starting from maltitol syrups with a maltitol content and a maltotriitol content in the ranges of those described in the invention, did not identify different maltitol crystals. The only given definition in terms of shape is "prism-like". Since they did not identify several types of crystals, of course they could not link that information to the maltotriitol content of the maltitol solution.

2. A few years later, Devos et al., in US 4,846,139, with a priority date of Dec. 1984, assigned to Roquette Frères, the Assignee of the present invention, described a process for the preparation of crystalline maltitol.

In this document, the maltitol syrup contains 87-97.5% of maltitol, less than 1% of DP \geq 4, and 2.5-13% of maltotriitol, see Col. 4 line 59 to Col. 5 line 7. The starting maltitol syrups thus have maltitol and maltotriitol contents in the range of those described in the invention.

The crystallization is also done out of water.

The crystals themselves are not discussed, and in particular their shape is not mentioned. Since no specific crystals are identified, the influence of the maltotriitol content on the shape of the obtained crystals was not mentioned or discussed either.

3. A decade later, Kawashima et al., in US 5,583,215 assigned to Towa, with an earliest priority date of June 1990, CIP filed in July 1994, described a new crystalline mixture solid containing maltitol (copy of patent enclosed).

The starting maltitol solution which is crystallized contains 80-98% maltitol, 0.5-15% sorbitol and 1.5-10% maltotriitol and DP \geq 3 (see Col. 4 lines 13-17). More specifically, the examples of this patent relate to maltitol solutions with the following compositions:

	Maltitol	Sorbitol	DP \geq 3
Ex.1	94.0	1.2	4.8
Ex.2	90.3	1.0	8.7
Ex.3	87.8	4.0	8.2
Ex.4	95.2	1.0	3.8

The crystallization is carried out by extrusion.

The obtained crystalline mixture is described as having a "*crushed and relatively tight crystal structure*" (Col. 3 lines 30-32). It is in fact clearly not pure maltitol, since its melting point is in the range 134°-145°C (Col. 3 line 41), compared to the melting point of maltitol which is 146.5 to 147°C.

Once again, even though the starting syrups have a maltitol content greater than 87%, and a maltotriitol content greater than 4%, or between 1 and 4%, i.e. in the ranges of those described in the present invention, the shape of the obtained crystals is not discussed. The qualitative and quantitative importance of the maltotriitol in terms of its consequence on the shape of the crystals has consequently not been determined.

4. A few years later, Caboche, in US 5,651,829, with a priority date of March 1995, assigned to Roquette Frères, the Assignee of the present invention, describes a novel crystalline maltitol composition.

This time, the crystallization is done by atomization.

The starting maltitol syrup contains at least 92% maltitol (Col. 6 lines 50-51), preferably at least 95%, better still at

least 98%, the ideal being close to or more than 99% (Col. 7 lines 11-14). The content of polyols other than maltitol should be low, preferably less than 5%, preferably still less than 2%. A long list of these other polyols is given, and maltotriitol is one of them (Col. 4 lines 45-50).

In the examples of this patent (see Table Col.10), the characteristics of the starting maltitol syrups are as follows:

Composition	Maltitol	Other polyols	Melting point
I1	99.8%	traces	149°C
I2	95.8%	2.9%	146°C

The starting maltitol syrups thus also have maltitol and maltotriitol contents in the range of those described in the instant invention.

In this patent, the obtained crystals are described, see Col.5 lines 15-24, as being *"porous and honeycombed"*, *"particles (...) essentially spherical, lacking sharp edges and composed of a vast number of crystalline micro-particles agglomerated to each other"*. Additionally, the observed structure is said to *"differ distinctly from that of a maltitol crystallized in water [Hirao et al., Devos et al.] and that of an extruded maltitol [Kawashima], which consist of very angular cubic or parallelepipedal particles"* (which incidentally confirms the *"prism-like"* description of Hirao).

Again, the inventor did not observe that maltitol could be obtained with different types of crystals, nor obviously did he make any link with the maltotriitol content of the starting syrup.

5. Kataura et al., in EP 741 140 filed by Towa, with an earliest priority of May 1995, describe a process for manufacturing crystalline maltitol, and the crystalline mixture solid containing crystalline maltitol.

Here, the crystallization is done out of water.

The starting maltitol syrup contains 92-99.9% maltitol (Col. 3 line 49). The syrup should contain little amounts of DP3 and DP6 (Col. 3 line 55). Again, the starting maltitol syrup has maltitol and maltotriitol contents in the range of those described in the present invention.

The shape of the crystalline maltitol is not commented upon, nor is the content of maltotriitol discussed.

Comments

From the first document relating to crystalline maltitol (in Feb.1981) until the invention (priority Sept. 1997), i.e. during more than 16 years, crystalline maltitol has been prepared by various methods (crystallization out of water, extrusion, atomization) starting from different compositions of maltitol syrups with a high maltitol content and presenting maltotriitol contents in the ranges of those described in the instant invention, i.e. either less than 1%, or at least 4%, or between 1 and 4%.

However, never during these 16 years were the two types of maltitol crystals observed, prismatic and bipyramidal.

The fact that a specific content (below 1%, between 1 and 4%, and at least 4%) of a specific polyol (maltotriitol) could

be used to selectively obtain one or both types of crystals has thus been ignored during this long period of time, which clearly demonstrates the non-obviousness of the present invention.

One should also point out that the Assignees of the patents discussed above (Hayashibara, Towa, Roquette Frères) are all well known and recognized worldwide leader producers of polyols and in particular maltitol.

It is therefore submitted that the process of the invention, i.e. the selection of the maltotriitol content in the starting maltitol syrup in order to obtain crystals of a specific shape, is definitely inventive, since these three expert firms, starting from the same maltitol syrups containing maltotriitol below 1%, at least 4% or between 1 and 4%, have all prepared maltitol crystals for over 16 years without ever noticing the different types of crystals nor determining the very specific parameter which directs the formation of each population, i.e. a specific content of a specific polyol.

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In view of the above, it is considered that the application is in proper form for allowance. Favorable consideration and prompt allowance of these claims are respectfully requested.

Respectfully submitted,

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